

# Quarter-filled spin density wave states with long-range Coulomb interaction\*

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Spin density wave (SDW) states at quarter-filling, which coexist with charge density wave (CDW) states, have been examined where the critical temperature is calculated for an extended Hubbard model with long range repulsive interactions. Within the mean-field theory, it is shown that the first order transition occurs with decreasing temperature for interactions located around the boundary between SDW state and CDW state.

KEYWORDS: A. Organic compound, D. Spin-density waves, D. Charge-density waves, D. Phase transition

## §1. Introduction

Bechgaard salts ( (TMTSF)<sub>2</sub>X and (TMTTF)<sub>2</sub>X ), which are known as low dimensional organic conductors, exhibit spin density wave (SDW) states at low temperatures.<sup>1,2)</sup> The SDW states exhibit several unconventional properties associated with charge density wave (CDW). The recent X-ray experiment on (TMTSF)<sub>2</sub>PF<sub>6</sub> salt<sup>3,4)</sup> has shown that 2k<sub>F</sub>-SDW coexists with 2k<sub>F</sub>-CDW at temperatures just below the onset temperature of SDW where k<sub>F</sub> denotes a Fermi momentum. The coexistence of SDW and CDW has been studied theoretically in terms of the mean-field theory at the absolute zero temperature. By taking into account repulsive interactions of both on-site and nearest-neighbor sites and dimerization, it has been demonstrated that 2k<sub>F</sub>-SDW coexists with 4k<sub>F</sub>-CDW.<sup>5)</sup> Further the coexistence of 2k<sub>F</sub>-SDW and 2k<sub>F</sub>-CDW has been found by adding the next-nearest-neighbor repulsive interaction.<sup>6)</sup>

In the present paper, by extending the previous calculations,<sup>5,6,7)</sup> we study if such a long range Coulomb interaction results in the coexistence even at the onset temperature of the SDW state, as found in the experiment.<sup>3,4)</sup>

## §2. Formulation

We examine a one-dimensional extended Hubbard model with interactions of on-site (*U*), nearest-neighbor (*V*), next-nearest-neighbor (*V*<sub>2</sub>) sites and dimerization energy (*t*<sub>d</sub>). The Hamiltonian is expressed as<sup>6)</sup>

$$H = - \sum_{\sigma=\uparrow,\downarrow} \sum_{j=1}^N (t - (-1)^j t_d) \left( C_{j\sigma}^\dagger C_{j+1,\sigma} + h.c. \right) + U \sum_{j=1}^N n_{j\uparrow} n_{j\downarrow} + \sum_{j=1}^N V_{1j} n_j n_{j+1} + V_2 \sum_{j=1}^N n_j n_{j+2} . \quad (1)$$

The quantity  $C_{j\sigma}^\dagger$  denotes the creation operator of the electron at the *j*-th site and  $V_{1j} = V - (-1)^j \delta V$  where  $\delta V$  also comes from dimerization and  $n_j = n_{j\uparrow} + n_{j\downarrow}$  with  $n_{j\sigma} = C_{j\sigma}^\dagger C_{j\sigma}$ . Quantities *t*, *k*<sub>B</sub> and lattice constant are taken as unity. Due to the quarter-filled band with the Fermi wave vector  $k_F = \pi/4$ , order parameters with  $m = 1, 2, 3$  are calculated self-consistently by

$$S_{mQ_0} = \frac{1}{N} \sum_{\sigma=\uparrow,\downarrow} \sum_{-\pi < k \leq \pi} \text{sgn}(\sigma) \left\langle C_{k\sigma}^\dagger C_{k+mQ_0,\sigma} \right\rangle_{\text{MF}} , \quad (2)$$

$$D_{mQ_0} = \frac{1}{N} \sum_{\sigma=\uparrow,\downarrow} \sum_{-\pi < k \leq \pi} \left\langle C_{k\sigma}^\dagger C_{k+mQ_0,\sigma} \right\rangle_{\text{MF}} , \quad (3)$$

where  $Q_0 = 2k_F$ ,  $S_0 = 0$ ,  $D_0 = 1/2$ ,  $S_{Q_0} = S_{3Q_0}^*$ ,  $D_{Q_0} = D_{3Q_0}^*$ ,  $S_{2Q_0} = S_{2Q_0}^* \equiv S_2$  and  $D_{2Q_0} = D_{2Q_0}^* \equiv D_2$ . In Eqs. (2) and (3),  $S_1 (\equiv |S_{Q_0}|)$ ,  $S_2$ ,  $D_1 (\equiv |D_{Q_0}|)$  and  $D_2$  correspond to the amplitudes for 2k<sub>F</sub>-SDW, 4k<sub>F</sub>-SDW, 2k<sub>F</sub>-CDW and 4k<sub>F</sub>-CDW respectively. From eqs. (2) and (3), the free energy per site with the quantity  $U/16 + V/4 + V_2/4$  subtracted is given by

$$F_{\text{MF}} = -\frac{T}{N} \sum_{\sigma} \sum_{0 < k \leq Q_0} \sum_{n=1}^4 \ln \left( 1 + \exp[(E_{n\sigma}(k) - \mu)/T] \right) + U \left[ -\frac{1}{8} - \frac{1}{2} (|D_{Q_0}|^2 - |S_{Q_0}|^2) - \frac{1}{4} (D_{2Q_0}^2 - S_{2Q_0}^2) \right] + V \left( -\frac{1}{2} + D_{2Q_0}^2 \right) + i\delta V (D_{Q_0}^2 - D_{Q_0}^{*2}) + V_2 \left( -\frac{1}{2} + 2|D_{Q_0}|^2 - D_{2Q_0}^2 \right) + \frac{\mu}{2} , \quad (4)$$

where *T* is temperature and  $\mu$  is a chemical potential determined by  $D_0 = 1/2$ . In eq. (4),  $E_{n\sigma}$  is the eigen value for the mean-field Hamiltonian,<sup>7)</sup>

$$H_{\text{MF}} = \sum_{\sigma=\uparrow,\downarrow} \sum_{-\pi < k \leq \pi} \left[ \left( \varepsilon_k + \frac{U}{4} + V + V_2 \right) C_{k\sigma}^\dagger C_{k\sigma} + \left( \Delta_{Q_0\sigma} C_{k+Q_0,\sigma}^\dagger C_{k\sigma} + h.c. \right) + \Delta_{2Q_0\sigma} C_{k\sigma}^\dagger C_{k+2Q_0,\sigma} \right] , \quad (5)$$

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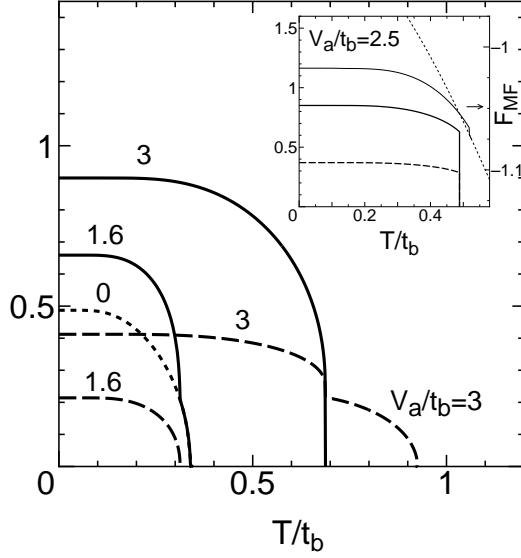


Fig. 1. The  $T/t_b$ -dependence of order parameters,  $S_1$  (solid and dotted curves) and  $D_2$  (dashed curve) for  $V_a/t_b = 0, 1.6$  and  $3$  with the fixed  $V_2/V_a = 0$ . In the inset, corresponding  $S_1$  and  $D_2$  for  $V_a/t_b = 2.5$  (left axis) is shown with free energies (right axis) for  $V_a/t_b = 2.5$  (thin solid curve) and the normal state (thin dotted curve).

where  $\varepsilon_k = -2t \cos k$ ,  $\Delta_{Q_0\sigma} = (U/2 - 2V_2)D_{Q_0} + 2i\delta V D_{Q_0}^* - \text{sgn}(\sigma)US_{Q_0}/2$  and  $\Delta_{2Q_0\sigma} = (U/2 - 2V + 2V_2)D_{2Q_0} - \text{sgn}(\sigma)US_{2Q_0}/2 - 2it_d \sin k$ . Here we note an excess free energy,  $\delta F$ , which is obtained by expanding  $F_{MF}$  in terms of order parameters. Since there are two kinds of coupling for  $D_2 S_1^2$  and  $S_1 S_2 D_1$  as found in the next section, the relevant expressions for  $\delta F$  are written as

$$\delta F = B_{S_1} S_1^2 + B_{D_2} D_2^2 + G_1 D_2 S_1^2 + \dots, \quad (6)$$

$$\delta F = B_{S_1} S_1^2 + B_{S_2} S_2^2 + B_{D_1} D_1^2 + G_2 S_1 S_2 D_1 + \dots, \quad (7)$$

where  $G_1$  and  $G_2$  are coupling constants. For small  $B_X$ , one obtains  $B_X \propto (T - T_X^0)$  with  $T_X^0$  ( $X = S_1, D_2$  and  $D_1$ ) being the onset temperature for  $S_1$ -state,  $D_2$ -state and  $D_1$ -state.

### §3. Phase diagram at finite temperatures

We examine SDW states at finite temperatures, by calculating eqs. (2), (3) and (4). The numerical calculation is performed by using  $t_b = t - t_d$  and  $V_a = V + \delta V$  with the fixed  $t_d/t = 1/21$ ,  $U/t = 3.81$  and  $\delta V/V = 1/9$ .<sup>6)</sup>

In Fig. 1,  $T$ -dependence of order parameters for  $V_2 = 0$  and  $V_a/t_b = 0, 1.6$  and  $3$  is shown where solid curve and dashed curve correspond to  $S_1$  and  $D_2$ , respectively and  $S_2 = D_1 = 0$ . The dotted curve with  $V_a = 0$  denotes a conventional  $2k_F$ -SDW state. There are two kinds of phase transitions for  $V_a/t_b = 1.6$  (3) where pure  $S_1$  state ( $D_2$  state) is obtained at high temperatures with  $0.314 < T/t_b < 0.341$  ( $0.688 < T/t_b < 0.924$ ) while a coexistent state of  $S_1$  and  $D_2$  is obtained at low temperature with  $T/t_b < 0.314$  ( $T/t_b < 0.688$ ). The inset denotes a first order transition into a coexistent state

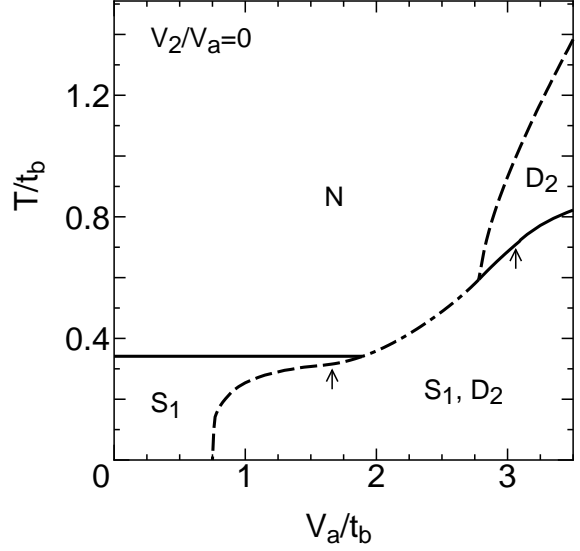


Fig. 2. Phase diagram on the plane of  $V_a/t_b$  and  $T/t_b$  with  $V_2/V_a = 0$  where  $S_1$ ,  $D_2$  and  $N$  correspond to  $2k_F$ -SDW,  $4k_F$ -CDW and normal state, respectively. In the interval region between two values ( $V_a/t_b$ ) shown by two arrows, a first order transition takes place with increasing temperature from the ground state.

of  $S_1$  and  $D_2$ , which occurs at  $T/t_b = 0.488$  for  $V_a/t_b = 2.5$ . The first order transition temperature is estimated by comparing  $F_{MF}$  of eq.(4). Based on these results, phase diagram on the plane of  $V_a/t_b$  and  $T/t_b$  for  $V_2 = 0$  is shown in Fig. 2, where  $S_1 \neq 0$  ( $D_2 \neq 0$ ) at temperatures below the solid curve (dashed curve). With decreasing temperature, the second order transition into  $S_1$  state occurs for  $V_a/t_b < 0.75$  while the second order transition into  $S_1$  state is followed by the second (first) order transition into the coexistent state of  $S_1$  and  $D_2$  for  $0.75 < V_a/t_b < 1.67$  ( $1.67 < V_a/t_b < 1.89$ ). It is noticeable that the first order transition from the normal ( $N$ ) state into the coexistent state of  $S_1$  and  $D_2$  takes place on the dash-dotted curve with  $1.89 < V_a/t_b < 2.78$ . Further the second order transition into  $D_2$  state is followed by the first (second) order transition into the coexistent state of  $S_1$  and  $D_2$  for  $2.78 < V_a/t_b < 3.07$  ( $3.07 < V_a/t_b$ ). Equation (6) indicates a fact that the first order transition originates in the third term with a coefficient  $G_1$  and that the second order transition from  $S_1$  state to the coexistent state of  $S_1$  and  $D_2$  is due to  $G_1 = 0$  in the pure  $S_1$  state. Thus the first order transition for  $V_2 = 0$  is attributable to the third term of eq. (6).

Next we examine another case of  $V_2/V_a = 1$ . In Fig. 3,  $T$ -dependence of order parameters is shown with some choices of  $V_a/t_b$  where the solid curve, dashed curve and dash-dotted curve correspond to  $S_1$ ,  $D_1$  and  $S_2$ , respectively. For  $V_a/t_b = 1.4$  (2.5), pure  $S_1$  state ( $D_1$  state) is obtained at high temperatures with  $0.223 < T/t_b < 0.341$  ( $0.746 < T/t_b < 1.00$ ) while a coexistent state of  $S_1$ ,  $D_1$  and  $S_2$  is obtained at low temperature with  $T/t_b < 0.223$  ( $T/t_b < 0.746$ ). The inset denotes a first order transition, which occurs at  $T/t_b = 0.4$  for  $V_a/t_b = 1.8$ . For  $V_2/V_a = 1.0$ , the phase diagram on the plane of  $V_a/t_b$  and  $T/t_b$  is shown in Fig. 4, where  $S_1 \neq 0$  ( $D_1 \neq 0$ ) below the solid curve (dashed curve). For

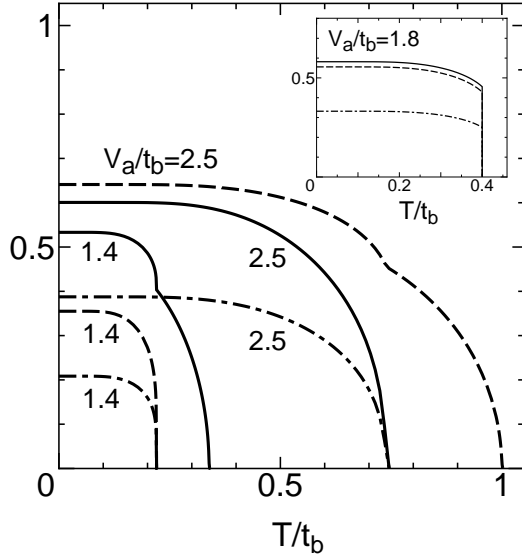


Fig. 3. The  $T/t_b$ -dependence of order parameters,  $S_1$  (solid curve),  $S_2$  (dash-dotted curve) and  $D_1$  (dashed curve) for  $V_a/t_b = 1.4$  and  $2.5$  with the fixed  $V_2/V_a = 1$ . The inset shows order parameters for  $V_a/t_b = 1.8$ , where  $S_1$ ,  $S_2$ , and  $D_1$  vanish followed by a jump at a critical temperature.

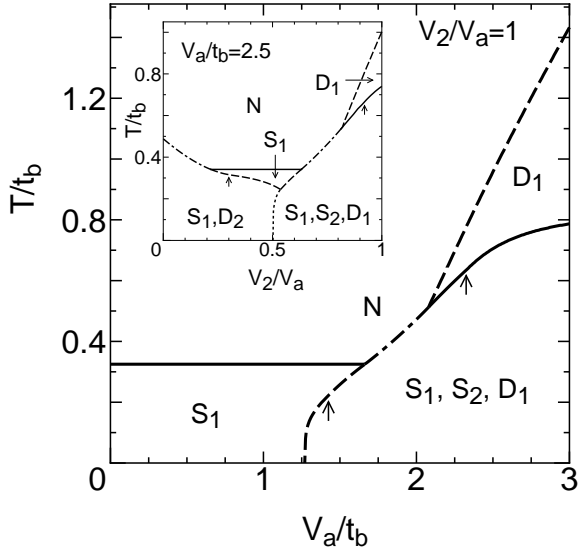


Fig. 4. Phase diagram on the plane of  $V_a/t_b$  and  $T/t_b$  with  $V_2/V_a = 1$ . The notations are the same as in Fig. 2, where  $S_2$  and  $D_1$  denote  $4k_F$ -SDW and  $2k_F$ -CDW, respectively. The inset denotes a phase diagram on the plane of  $V_2/V_a$  and  $T/t_b$  with  $V_a/t_b = 2.5$ .

$V_a/t_b < 1.26$ , the second order transition into  $S_1$  state occurs while the second order transition into  $S_1$  state is followed by the second (first) order transition into the coexistent state of  $S_1$ ,  $D_1$  and  $S_2$  for  $1.26 < V_a/t_b < 1.43$  ( $1.43 < V_a/t_b < 1.66$ ). A salient feature is the first order transition on the dash-dotted curve, where the normal ( $N$ ) state moves into the coexistent state of  $S_1$ ,  $D_1$  and  $S_2$  in the interval region of  $1.66 < V_a/t_b < 2.08$ . The second order transition into  $D_1$  state is followed by the first (second) order transition into the coexistent state of  $S_1$ ,  $D_1$  and  $S_2$  for  $2.08 < V_a/t_b < 2.32$  ( $2.32 < V_a/t_b$

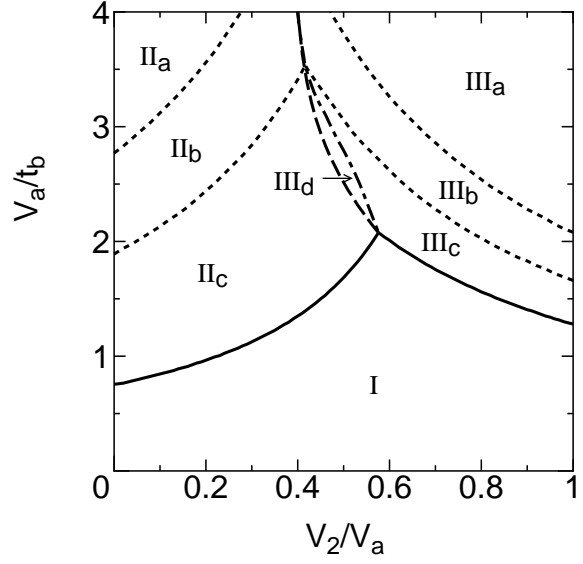


Fig. 5. Phase diagram on the plane of  $V_2/V_a$  and  $V_a/t_b$ . The solid curve (dashed curve) shows the boundary for  $T = 0$ , between the pure  $S_1$  state and the coexistent state (between two kinds of coexistent state).<sup>7)</sup> The dotted curve corresponds to a boundary at the critical temperature where it is obtained from the intersection of the solid curve and the dashed curve in Figs. 2 and 4. The respective regions together with the dash-dotted curve are depicted in the main text.

). The first order transition originates in the third term with a coefficient  $G_2$  of eq. (7). It should be noticed that the existence of  $S_2$  is crucial to obtain a direct transition from  $N$  state to the coexistent of  $S_1$  and  $D_1$ . Actually, for  $V_a/t_b = 2.5$  and  $V_2/V_a = 0.7$ , the mean-field calculation with  $S_2 = 0$  leads to a second order transition from  $N$  state to pure  $S_1$  state,<sup>8)</sup> while the corresponding calculation in the presence of  $S_2$  exhibits a first order transition into a coexistent state.

Base on these results, a phase diagram on a plane of  $V_2/V_a$  and  $V_a/t_b$  is shown in Fig. 5. With decreasing temperatures, we obtain the following phase transitions. In the region I, a transition from  $N$  state into pure  $S_1$  state appears while the region II<sub>a</sub> (II<sub>c</sub>) shows the successive transition given by  $N \rightarrow D_2 \rightarrow S_1$  and  $D_2$  ( $N \rightarrow S_1 \rightarrow S_1$  and  $D_2$ ). The first order transition from  $N$  state into a coexistent state of  $S_1$  and  $D_2$  is obtained in the region II<sub>b</sub>. In the region III<sub>a</sub> (III<sub>c</sub>), there is the successive transition given by  $N \rightarrow D_1 \rightarrow S_1$ ,  $D_1$  and  $S_2$  ( $N \rightarrow S_1 \rightarrow S_1$ ,  $D_1$  and  $S_2$ ). The first order transition from  $N$  state into a coexistent state of  $S_1$ ,  $D_1$  and  $S_2$  is obtained in the region III<sub>b</sub> while the region III<sub>d</sub> corresponds to the transition given by  $N \rightarrow S_1 \rightarrow S_1, D_2 \rightarrow S_1, D_1$  and  $S_2$ . The region III<sub>d</sub> is understood by an example shown in the inset of Fig. 4 on the plane of  $V_2/V_a$  and  $T/t_b$ , where the corresponding transition is obtained for  $0.5 < V_2/V_a < 0.54$  for  $V_a/t_b = 2.5$ .

By studying a model at quarter-filling with long range Coulomb interactions, we have found that, with decreasing temperature, a first order transition from  $N$  state into the coexistent state of SDW and CDW occurs. Since there is a reasonable range of parameters, it is considered that the present result of the first order transition could

be relevant to the coexistent state of  $2k_F$ -SDW and  $2k_F$ -CDW found in the X-ray experiment.<sup>3,4)</sup>

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